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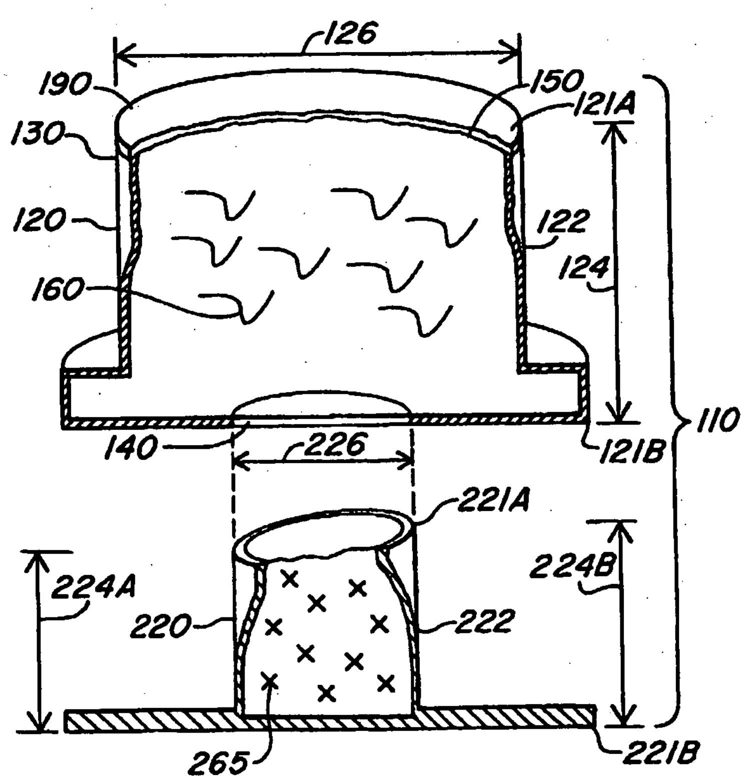

(57) Abstract: Disclosed are apparatus for delivery of a gaS (10), e.g., carbon dioxide and/or chlorine dioxide, and methods of its use and manufacture. The apparatus includes a vessel (20) defining a reaction volume, the vessel including a sachet layer (50) disposed about an aperture (30) defined by the vessel, and a puncturable surface (40). The vessel contains one or more reactants (60) that generate a gas in the presence of an initiating agent, e.g., water. The apparatus can include a second vessel (220) formed to engage the puncturable surface to deliver initiating agent and/or a second reactant to the vessel (20).

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APPARATUS AND METHODS FOR DELIVERY OF A GAS

Related Applications

This application claims priority to pending U.S. Provisional Patent Application Serial No. 60/341,429, filed December 17, 2001, and pending U.S. Provisional Patent Application Serial No. 60/405,202, filed August 22, 2002, the entire disclosures of which are hereby incorporated by reference herein.

Field of the Invention

The present invention relates generally to apparatus and methods for delivery of gas, and more specifically, to apparatus and methods for initiating gas generation and controlling the amount, rate and duration of gas delivery.

Background of the Invention

The use of gas for retarding controlling, killing and preventing microbiological contamination (*e.g.*, bacterial, fungi, viruses, mold spores, algae, and protozoa); retarding, preventing, or controlling biochemical decomposition; controlling respiration, deodorizing and/or retarding and preventing chemotaxis to name a few, is known. Such gases include, but are not limited to, chlorine dioxide, sulfur dioxide, nitrogen dioxide, nitric oxide, carbon dioxide, hydrogen sulfide, hydrocyanic acid, and dichlorine monoxide.

In particular, chlorine dioxide has been found to be useful as a disinfectant antiseptic and sanitizer. It is used, *e.g.*, to disinfect drinking water and various water supplies. In addition, chlorine dioxide finds use as a bleaching agent for flour, fats, and textiles. Chlorine dioxide also has shown great utility as an antiseptic for treating metal and plastic surfaces, as well as other substrates such as countertops, meat processing and packaging equipment, and dental and medical instruments and devices.

One disadvantage of the prior art methods for generating chlorine dioxide generally is that unsatisfactory levels of by-products or reactants remain as a residue. For example, in the case of chlorine dioxide gas, the byproduct chlorite leaves residues on food handling equipment and medical and dental surfaces. Human contact with such residues should be avoided or substantially minimized according to FDA and EPA regulations.

Another requirement in the food handling and related industries is the need for raw materials or ingredients that are safe to handle in the preparation of the disinfectant. The requirement is for the inclusion of reagents that are safe to use and, upon generating chlorine dioxide, produce byproducts that are non-toxic and/or biodegradable.

Moreover, chlorine dioxide cannot be transported commercially as a concentrated gas for safety reasons, and instead has been generated at the site where it is used. On-site generation plants used to generate gas are expensive and take up valuable space. Moreover, even when prior art apparatus do not require a separate gas generation plant, such apparatus are undesirable because the amount of gas generated, the efficiency of the generation, and the duration of the gas generation is neither controlled nor optimized.

Summary of the Invention

A novel approach to the generation and delivery of a gas has now been discovered. The present invention includes a unique delivery system that controls the rate and efficiency of gas-producing reactions. Moreover, by using discreet amounts of reactant contained with a defined reaction volume, the skilled practitioner can fabricate a gas delivery apparatus that is compact, cost-effective, and safe. Furthermore, the present invention can be used for a variety of applications, including delivery of gas to air or water, for a variety of purposes including disinfection, deodorization, bleaching and sanitization.

In one embodiment, the present invention features an apparatus for delivery of a gas that generally includes a vessel defining a reaction volume, and a reactant disposed within the vessel that generates a gas in the presence of an initiating agent. The vessel includes a sachet layer disposed about an aperture defined by the vessel, and a puncturable surface. In one embodiment, the reactant includes an aqueous soluble acid and an aqueous soluble chlorite. In other embodiment, the sachet layer comprises a hydrophobic material. In a preferred embodiment, the sachet layer comprises a water vapor selective film.

Apparatus of the invention can further include an envelope layer disposed about the aperture and disposed adjacent to the sachet layer. In certain embodiments, the sachet layer is constructed from a hydrophilic material and the envelope layer is constructed from a hydrophobic material. The apparatus can include a sachet disposed within the vessel, wherein the reactant is disposed within the sachet. Such apparatus can further include a second reactant disposed outside of the sachet and within the vessel. The puncturable surface can be constructed from a retractable material or a frangible material.

In yet another embodiment, the apparatus further includes a second vessel formed to engage the vessel and to provide a compound. The compound can include a second reactant and/or an initiating agent. The second vessel can be a needle. Alternatively, the second vessel can be formed to couple with the vessel and further define the reaction volume. In

such embodiments, the second vessel can include a male member that is formed to slidably engage the vessel upon actuation of the male member. The second vessel and the vessel can define complementary surfaces for a snap fit upon actuation of the male member. In another embodiment, the second vessel and the vessel define complementary threads for threaded engagement.

In another aspect, the invention features a kit for delivery of a gas. The kit generally includes a first vessel defining a reaction volume, the first vessel including a sachet layer disposed about an aperture defined by the vessel, and a puncturable surface. The kit also includes a reactant disposed within the first vessel that generates a gas in the presence of an initiating agent and a second vessel selectively engageable with the first vessel and formed to provide a compound. In one embodiment, the reactant includes an aqueous soluble acid and an aqueous soluble chlorite. In other embodiment, the sachet layer comprises a hydrophobic material. In a preferred embodiment, the sachet layer comprises a water vapor selective film.

In one embodiment, the vessel can further include an envelope layer disposed about the aperture and disposed adjacent to the sachet layer. In certain embodiments, the sachet layer in constructed from a hydrophilic material and the envelope layer is constructed from a hydrophobic material. The apparatus can include a sachet disposed within the vessel, wherein the reactant is disposed within the sachet. Such apparatus can further include a second reactant disposed outside of the sachet and within the vessel. The puncturable surface can be constructed from a retractable material or a frangible material.

In yet another embodiment, the kit further includes a second vessel formed to engage the vessel and to provide a compound. The compound can include a second reactant and/or an initiating agent. The second vessel can be a needle. Alternatively, the second vessel can be formed to couple with the vessel and further define the reaction volume. In such embodiments, the second vessel can include a male member that is formed to slidably engage the vessel upon actuation of the male member. The second vessel and the vessel can define complementary surfaces for a snap fit upon actuation of the male member. In another embodiment, the second vessel and the vessel define complementary threads for threaded engagement.

In yet another embodiment, the present invention features a method of delivering a gas. The method includes the step of providing an apparatus for delivery of a gas including a vessel defining a reaction volume. The vessel generally includes a sachet layer disposed

about an aperture defined by the vessel, and a puncturable surface. Reactant is disposed within the vessel that generates a gas in the presence of an initiating agent. This embodiment further includes the step of delivering an initiating agent to the vessel to initiate delivery of the gas. In one embodiment, the method features the further steps providing a second vessel formed to engage the vessel and deliver a compound (*e.g.*, initiating agent and/or reactant), and engaging the vessel and the second vessel to deliver the compound.

In yet another embodiment, the present invention features a method of delivering a gas. The method generally includes providing a vessel including an aperture defined by the vessel and a puncturable surface, disposing a reactant within the vessel that generates a gas in the presence of an initiating agent, and sealing a sachet layer about the aperture defined by the vessel to define a reaction volume.

The invention will be understood further upon consideration of the following drawings, descriptions and claims.

Description of the Drawings

The invention is pointed out with particularity in the appended claims. The drawings are not necessarily to scale, with emphasis instead generally being placed upon illustrating the principles of the invention. The advantages of the invention can be better understood by reference to the description taken in conjunction with the accompanying drawings, in which:

Figure 1 illustrates an exemplary embodiment of an apparatus for delivery of a gas in accordance with the present invention.

Figures 2A and 2B illustrate further exemplary embodiments of apparatus for delivery of a gas that include second vessels that are formed for pressure fit and threaded engagement, respectively.

Figures 3A and 3B further illustrate the embodiments depicted in Figures 2A and 2B, respectively, where the second vessel is engaged with the first vessel.

Like reference characters in the respective drawn figures indicate corresponding parts.

Detailed Description of the Invention

A novel approach to the delivery of gas has now been discovered. By using a vessel defining a reaction volume, gas can be generated efficiently and safely. By using a vessel having a puncturable surface, the user may initiate gas generation by delivering reactant and/or initiating agent via the puncturable surface. Moreover, by using a vessel that includes a sachet layer disposed about an aperture defined by the vessel, the reaction can be controlled

by controlling the release of gas through the sachet layer. Upon initiation, the reactant generates the gas that exits the apparatus through the sachet layer. The gas delivery apparatus is compact, cost-effective and safe. The present invention can be used for a variety of applications, including delivery of gas to air or water, and for a variety of purposes including disinfection, deodorization, decontamination, bleaching and sanitization.

One advantage to this approach is that at user demand, the user can initiate the reaction that generates the gas. Another advantage is that the reactants, which can be dangerous to handle directly, are isolated from contact with the user because the reactant is enclosed within the vessel until the reaction is initiated. Yet, another advantage is that the reactants and any by-products remain enclosed in the apparatus before, during and after the reaction is initiated.

Another advantage is that the apparatus of the present invention do not allow for the dilution of the reactant. Because the reactant remains concentrated within the reaction volume, less reactant is necessary to drive the reaction to completion, and the reaction is more efficient than it would be if the reactants were diluted. Furthermore, because the reaction is driven to completion, unreacted reactant is minimized or eliminated. The reactant concentration also minimizes unwanted by-products.

Yet another advantage is that the apparatus is small and stable, and thus can be easily and economically shipped, stored, and administered. Still a further advantage is that the apparatus can be manipulated to allow for either rapid or slow delivery of gas, *e.g.*, by selection of sachet layer material, and optional envelope layer material. Another advantage is that the apparatus can be designed to deliver gas to either a gaseous environment, *e.g.*, ambient air, or a liquid environment, *e.g.*, water. Other advantages will be evident to the practitioner having ordinary skill in the art.

In order to more clearly and concisely describe the subject matter of the claims, the following definitions are intended to provide guidance as to the meaning of specific terms used in the following written description, examples and appended claims.

As used herein, the term "vessel" refers to a hollow receptacle defining a reaction volume. The vessel defines an aperture, *e.g.*, an opening positioned on the vessel. The vessel generally includes a puncturable surface for the introduction of a compound (*e.g.*, initiating agent or reactant), and a sachet layer disposed about an aperture defined by the vessel.

"Reactant" refers to a reactant or a mixture of reactants that generate gas in the presence of an initiating agent. Such reactants include, but are not limited to metal chlorites (*e.g.,* sodium chlorite), and acids (*e.g.,* citric acid). The reactant or reactants can further include additives, such as activated hydrotalcite. As used herein "initiating agent" refers to any agent that initiates the generation of gas from the reactant. For purposes of the present invention, initiating agent includes, but is not limited to, gaseous or liquid water. The initiating agent can enter the apparatus through the sachet layer and/or the initiating agent can be included within the apparatus, *e.g.*, contained in a second vessel that is coupled with the first vessel.

"Puncturable surface" refers a surface that may be pierced or punctured when a force is applied to the surface. A puncturable surface may comprise material that retains a puncture hole where it was punctured. Alternatively, a puncturable surface may comprise material that retracts or contracts to an unpierced (*e.g.*, a closed) configuration after being punctured (*e.g.*, by a needle). Another example of a puncturable surface is a frangible surface that ruptures upon application of pressure, *e.g.*, by actuation of a second vessel against the puncturable surface, thereby allowing the second vessel to engage the first vessel.

The term "sachet layers" refers to layers that allow exit of the gas from the apparatus through the sachet layer. Sachet layers typically are constructed from a planar material, such as, but not limited to, a polymeric sheet or film. Sachet layers include any of the layers described herein and in pending United States Patent Application Serial No. 10/225,769, filed August 22, 2002, and incorporated herein by reference.

The term "sachet" refers a closed receptacle for reactant. The sachet is "closed" in the sense that the reactants are substantially retained within the sachet and the sachet volume is substantially sealed around its perimeter. The sachets of the present invention also can include further materials, *e.g.* a sachet can comprise a barrier layer and a sachet layer sealed about the perimeters of the layers to define a closed receptacle for reactant. Another example of a sachet is a rigid frame defining one or more openings and one or more layers, including at least one sachet layer, disposed about one or more openings to define a closed receptacle for reactant. Further examples and embodiments are described in greater detail herein.

"Envelope layers" are layers that allow exit of the gas from the apparatus through the envelope layer, and are disposed adjacent to at least one sachet layer such that the sachet layer is on the reaction volume side of envelope layer. Envelope layers typically comprise a

planar material such as a sheet or film, including, but not limited to, perforated films, non-perforated films and membranes. Further materials that can be used to construct envelope layers are described in pending United States Patent Application Serial No. 10/225,769, filed August 22, 2002. Relying upon the teaching disclosed herein and in pending United States Patent Application Serial No. 10/225,769, filed August 22, 2002, and the general knowledge in the art, the practitioner of ordinary skill will require only routine experimentation to identify one or more envelope or sachet layers and/or construct one or more envelopes or sachets adapted for the purposes at hand.

The term "envelope" means a closed receptacle wherein the envelope volume is sealed substantially about its perimeter, and contains at least one sachet and allows release of the gas from the envelope.

"Impermeable layer," as used herein, refers to a layer that substantially prevents or hinders passage of initiating agent. As contemplated herein, the impermeable layer does not participate in the generation of gas in that it does not facilitate contact between initiating agent and reactant. Impermeable layers can be constructed from various materials, including polymeric material, glass, metal, metallized polymeric material and/or coated papers. As used herein, barrier layers are impermeable layers. Preferred materials for impermeable layers and barrier layers may be any of the materials described in greater detail throughout this application and in pending United States Patent Application Serial No. 10/225,769, filed August 22, 2002.

The term "dry application" for the purposes of this application means at least an application where the apparatus of the present invention is not immersed in water or any other liquid. The term "wet application" for the purposes of the present invention means at least an application where the apparatus of the present invention is immersed in water, or other liquid, which can optionally include water.

Generation of a gas, *e.g.*, by acid activation, is well known in the art. For example, chlorine dioxide (ClO_2) is generated from sodium chlorite and an acid, such as citric acid, in the presence of moisture. Alternatively, chlorine dioxide can be produced by the reduction of a chlorate, *e.g.*, sodium chlorate or potassium chlorate, in the presence of an acid, *e.g.*, oxalic acid. Another example of generation of a gas by acid activation is the activation of a sulfite, *e.g.*, sodium bisulfite or potassium bisulfite, with an acid, *e.g.*, fumaric acid and/or potassium

bitartrate, in the presence of moisture to form sulfur dioxide. Yet another example is the acid activation of a carbonate, *e.g.*, calcium carbonate with an acid, *e.g.*, citric acid, to form carbon dioxide. Other applications will be apparent to the skilled practitioner. For example, the generation of nitrogen dioxide by the acid activation of a nitrite, *e.g.*, sodium nitrite or potassium nitrite. Alternative routes for generation of a gas, *e.g.*, reduction of chlorates by sulfur dioxide (Mathieson Process) are well known in the art and can be utilized in accordance with the present invention.

The present invention relates to apparatus and methods for delivering biocidal-effective amounts of a gas such as chlorine dioxide and sulfur dioxide. Chlorine dioxide gas may be released into a volume of water or other fluid to make a solution of the gas. The apparatus and methods of the present invention achieve delivery of a desired amount of gas, at a desired rate of generation, at a desired release rate, over a desired time period. This is accomplished by disposing suitable reactants in a defined and confined reaction volume such that upon initiation, by the user, the reactants, initiating agent, products, and by-products are held within a desired concentration range. The amount, rate and duration of delivery can be manipulated by, *e.g.*, choice of sachet layers, envelope layers, reactant amount, reactant ratio, aperture size, aperture shape, aperture location, reaction volume, and sachets and envelopes disposed within the vessel. Such manipulations can be exercised by the artisan using only routine experimentation in view of the teachings disclosed herein, described in pending United States Patent Application Serial No. 10/225,769, filed August 22, 2002, together with knowledge in the art.

The present invention is further illustrated by the following exemplary embodiments that should not be construed as limiting.

Figure 1 illustrates a perspective view of an exemplary embodiment of an apparatus 10 for delivery of a gas constructed in accordance with the present invention. In a general overview, apparatus 10 includes a vessel 20 that includes a sachet layer 50 disposed about aperture 30, and has a puncturable surface 40. The sachet layer 50 is substantially sealed about the periphery of the aperture 30 such that aclosed reaction volume is defined within the vessel. A reactant 60 is disposed inside the vessel 20 that generates a gas in the presence of an initiating agent. The sachet layer 50 allows exit of the gas through the sachet layer. The apparatus 10 may optionally contain a sachet 90 which contains a second reactant 92.

Apparatus 10 is particularly useful for user-initiated reaction, generation and delivery of a gas. For example, puncturable surface 40 can be utilized to provide initiating agent 80 to initiate reaction generation with reactant 60 with a syringe 75. After the initiating agent is introduced into the vessel, the apparatus generates gas in a controlled manner. In one embodiment, the apparatus provides a predetermined concentration of gas, which is released through the sachet layer from the reaction volume in a controlled and/or sustained concentration and/or rate.

Vessels of the invention may be fabricated from various materials and the materials may be, for example, impermeable layer materials or barrier layer materials. Suitable materials for impermeable layers and barrier layers may be any of the materials described in greater detail in this patent application and in pending United States Patent Application Serial No. 10/225,769, filed August 22, 2002. The barrier layer materials may have a rigid or semi-rigid structure. Suitable materials that may be used to fabricate vessels include, *e.g.*, metals, polymeric materials, metallized polymeric materials, and glass.

The vessel can be shaped and sized to suit a particular gas delivery application. For example, the vessel may be shaped so that it can be inserted into another device, *e.g.*, a cartridge, a humidifier, a vaporizer, a pipe or other machine. The vessel also can be formed into a shape such that the apparatus is easily inserted into or removed from various environments. For example, when sanitizing a pipe system, the user may initiate gas generation in a cylindrical vessel adapted to fit snugly with the inner diameter of a pipe, cartridge, or pipe system for transporting fluid. Alternatively, a cylindrical vessel size may be adapted to move through the inner diameter of a pipe system along with the fluid traveling through the pipe.

Suitable vessel geometries include cylindrical, oval, spherical, rectangular, and pyramid. Vessels can be formed into various geometries by various manufacturing methods, including, *e.g.*, the manufacturing methods describing forming barrier layer material into various geometries as described in pending United States Patent Application Serial No. 10/225,769, filed August 22, 2002.

Referring to Figure 1, the vessel 20 may be a cylinder having a first end 21A and a second end 21B. Vessel wall 22 preferably has a thickness suited to the materials of construction, the reactant and/or the space into which vessel 20 will be inserted. The vessel height 24 and the vessel diameter 26 also may be dimensioned to suit a particular application.

The aperture of the vessels of the present invention may be sized and/or shaped to deliver a quantity of gas suited to the intended application. Suitable aperture geometries include spheres, circles, ovals, triangles, rectangles, squares, and slits. For example, the vessel can define one or multiple apertures having similar or dissimilar shapes for the introduction of one or more compounds. The size, shape, and/or location on vessel where the one or more apertures are disposed may be selected to control, *e.g.*, limit or maximize, the quantity of gas released from the vessel. Apertures can be excised from or otherwise defined in the vessel after the vessel is formed or, alternatively, apertures may be molded or formed into the vessel during its manufacture. In one embodiment, the vessel aperture is formed to include an edge about its perimeter for forming a seal with sachet layer and/or an envelope layer or other additional layers.

The sachet layers of the present invention can be chosen to control the diffusion of the reactants out of the sachet, control the rate of gas release from the reaction volume and control the initiation of the reactants. For example, a hydrophilic sachet layer can increase the rate at which water and/or water vapor diffuses into the sachet; and the pore size and thickness of the sachet layer can effect the passage of water, reactants and gas through the sachet layer. Preferably the sachet layers include materials that are durable and stable during construction, storage and reaction, and are capable of fusing upon the application of heat or ultrasonic energy for construction purposes, *e.g.*, so that a sachet layer can be fused about its perimeter to an aperture to construct a vessel, or another sachet layer to construct a sachet.

The sachet layers of the present invention can be constructed of various materials, including polymeric material or coated papers, selective transmission films, woven material, non-woven membrane, membranes including extruded membranes, or perforated films. Also suitable for use in constructing the sachet layers of the present invention are composite layers, including, but not limited to, starch/polymer composite layers. Additionally, the sachet layer of the present invention can be constructed from material that is hydrophobic and/or hydrophilic. For example, the sachet layer can be constructed from a material having one or more hydrophilic zones and one or more hydrophobic zones. These zones can be created, *e.g.*, by printing a functional chemical group or polymer onto a surface of the sachet that is hydrophilic, hydrophobic, or charged to create one or more hydrophilic, hydrophobic, or charged zones. Yet another alternative embodiment uses a material to construct the sachet

layer that has a first surface that is hydrophilic and a second surface that is hydrophobic, formed, *e.g.*, by coextrusion or coating.

Preferably, the sachet includes a material that is water vapor selective. The term "water vapor selective" as used herein refers to a material that selectively allows permeation of water vapor and substantially impedes permeation of liquid water. More preferably, the material excludes permeation of liquid water. Typically, the water vapor selective material is hydrophobic. The skilled practitioner typically refers to water vapor selective material as water impermeable, although water vapor can permeate the layer, and refers to materials that allow permeation of liquid water as water permeable. Suitable water vapor selective materials can be made from a variety of materials including, but not limited to, polytetrafluoroethylene (PTFE), polypropylene (PP), polyethylene (PE), and fluorinated ethylene propylene (FEP).

Water vapor selective material is particularly advantageous because it substantially impedes or excludes the diffusion of water soluble species, such as water soluble reactants, additives, and reaction by-products, out of the apparatus. Membranes that allow liquid water permeation allow soluble species such as soluble reactants to permeate the apparatus and enter the environment. Prior attempts have been made to ameliorate or avoid escape of soluble species by using insoluble reactants or by binding or otherwise disposing the soluble species on insoluble materials such as clays and molecular sieves. This is not advantageous because it introduces additional steps and/or expense to the preparation of the apparatus. Thus, the water vapor selective materials of the present invention are advantageous because they impede or preclude the permeation of the soluble reactants, additives, and by-products, thus removing the need to include or generate insoluble reactants, by-products and/or additives.

Moreover, water vapor selective materials are advantageous because their use eliminates the need to use any further layers, *e.g.*, further sachet and/or envelope layers, to retain soluble species in the apparatus, which introduces additional steps and expense in the construction of the apparatus.

Preferably, the water vapor selective material has a thickness between about 5 microns and about 400 microns thick, a pore size between about 0.05 microns and about 10 microns, and a water intrusion pressure between about 30 millibars and about 4,000 millibars. Preferably the water vapor selective material is adhered to or otherwise supported by a

support layer that allows liquid water to permeate the support layer, and the overall thickness of both the water vapor selective layer and the support layer is between about 1 mil and 20 mils. More preferred, are water vapor selective materials having a thickness between about 15 microns and about 200 microns thick, a pore size between about 0.25 microns and about 5 microns, and a water intrusion pressure between about 100 millibars and about 1,500 millibars. Preferably, this layer has a water permeable support layer such that the total thickness of both layers is between about 2 mils and about 10 mils. Most preferred, are water vapor selective materials having a thickness between about 20 microns and about 100 microns thick, a pore size between about 1 micron and about 3 microns, and a water intrusion pressure between about 200 millibars and about 750 millibars. Preferably this layer has a water permeable support layer such that the total thickness of both layers is between about 3 mils and about 8 mils.

Generally, if rapid generation and release of gas is desired, a thinner layer is preferred because the thinner the layer, the more rapidly water vapor can diffuse into the apparatus and initiate the reaction, and the more rapidly gas can diffuse out of the apparatus. Conversely, if slower generation and release of gas is desired a thicker material is preferred, such as between about 5 mils and about 20 mils. Additionally or alternatively, if slower generation and release of gas is desired, a relatively smaller surface area of the material can be used.

In addition or alternatively, pore size of the water vapor selective layer can be selected to produce the desired release at specific depths of water in which the apparatus will be used. A smaller pore size will correspond to deeper operation by increasing the amount of hydraulic water pressure that the membrane will experience while remaining impermeable to liquid water.

Water vapor selective layers suitable for use in constructing the apparatus of the present invention preferably have water vapor permeability of between about 2,000 $g/m^2/24hrs$ and about 150,000 $g/m^2/24hrs$, as determined by JIS L 1099-1985 (Method B), "Testing Methods for Water Vapour Permeability of Clothes," from the Japanese Standards Association. Water vapor selective layers preferably have a resistance to liquid water permeation of at least about 30 millibars as determined by ISO 811-1981 "Textile fabrics - Determination of resistance to water penetration - Hydrostatic pressure test" published by the International Organization for Standardization.

Optionally, the water vapor selective layer or layers of the present invention can include a support layer to increase the strength of the layer, and/or to increase its ability to bond to the other materials used to construct the apparatus. The support layer preferably allows diffusion or passage of initiating agent to the surface of the water vapor selective layer. For example, the support layer can be spun, perforated or have large pores that allow passage of liquid water and vapor to the surface of the water vapor selective layer. The support layer can be affixed to the water vapor selective layer by any means, for example, lamination, casting, co-extrusion, and/or adhesive layers. Preferably, the apparatus is constructed so that the water vapor selective layer faces the interior of the vessel. The support layer itself can be hydrophilic and/or hydrophobic. If hydrophilic, the material can be used to attract and deliver liquid water and/or vapor to the surface of the water vapor selective material. Suitable support layers include, but are not limited to, polyethylene, polypropylene, nylon, acrylic, fiberglass, and polyester in the form of woven, non-woven, and mesh layers. Preferably, the support layer thickness is between about 1 mil and 20 mils.

Suitable water vapor selective materials include the 0.60 pore size, hydrophobic, polypropylene (PP) membrane having a thickness between about 250 microns and about 300 microns sold under the designation 060P1 by Cuno Incorporated (Meriden, CT). Also suitable is the 0.65 micron pore size, hydrophobic polyethylene material sold under the trade designation DOHP by Millipore (Bedford, MA). Another water vapor selective material suitable for use in a rapid release apparatus, includes a 1.75 mil thick, hydrophobic polytetrafluoroethylene (PTFE) layer thermally bonded to a 5 mil thick, hydrophobic polyethylene (PE) support layer sold under the trade designation BHA-TEX® by BHA Technologies (Kansas City, MO). Its resistance to liquid water permeation is at least about 500 millibar.

In embodiments where the sachet layer enables the initiating agent, *e.g.*, water and/or water vapor, to diffuse into the vessel, a protective seal made from impermeable materials can be placed over the sachet layer to prevent initiation until the user removes the protective seal. The user may remove the protective seal by, *e.g.*, peeling the protective seal off of the sachet layer prior to or after initiating the reaction.

The reactant contained within the vessel, and optionally, a second vessel as described below, can be in solid or solubilized form, *e.g.*, aqueous soluble reactant can be solubilized in water. Reactant can be maintained loose within the vessel and/or in one more sachets

disposed in the vessel. For example, citric acid can be maintained in a sachet disposed in a vessel and sodium chlorite can be maintained loose in the vessel exterior to the sachet. Additives can be included in the reactant as well.

The puncturable surface of the present invention is utilized to deliver a compound to the vessel, typically, a reactant and/or initiating agent that will commence reaction. Thus, the puncturable surface is constructed to suit the means of delivery of the compound. For example, the puncturable surface can be retractable such that the compound can be injected and the integrity of the vessel maintained after removal of the injection device. In another embodiment, the puncturable surface is frangible such that it is ruptured upon application of pressure. This embodiment can be used when engaging or coupling a second vessel to the first vessel, so that the barrier to delivery of the compound in the second vessel is removed by application of pressure. It yet another embodiment, the puncturable surface can be a removable layer which can be removed or peeled aside allowing the administration of a second compound or engagement of a second vessel. Optionally, the puncturable surface can be removed and replaced after delivery of a compound, *e.g.*, by threaded engagement, pressure fit, or snap fit with the vessel.

Preferably, the puncturable surface portion of vessel is constructed from impermeable or barrier layer material to prevent premature initiation. The puncturable surface can be manufactured from, *e.g.*, a polymer or a rubber material. Suitable puncturable surface materials include, for example, polyethylene, polypropylene, polystyrene, polyvinyledene chloride, polyvinyl chloride, latex rubber and/or other polymers. In one embodiment, a protective seal can be placed over the puncturable surface. The protective seal may protect the puncturable surface from inadvertently being pierced. The user removes the protective seal, for example, by peeling it from the vessel, prior to initiating the reaction by, *e.g.*, pushing the second vessel into the puncturable surface.

In one embodiment, for example as shown in Figure 1, the user employs a needle 70 (*e.g.*, a hypodermic needle), to pierce the puncturable surface 40, then plunges the syringe 75 to release a compound 80 (*e.g.*, initiating agent and/or additional reactants), into vessel 20. By releasing initiating agent into the vessel and/or providing further reactants, the user can initiate a gas generating reaction within the apparatus. Gas is then delivered to the atmosphere surrounding apparatus through the sachet layer in a controlled, safe, and efficient manner.

Figure 2A is a cross-sectional side view of another embodiment of an apparatus 110 for delivery of a gas constructed in accordance with the present invention. In general overview, apparatus 110 includes a vessel 120, which defines an aperture 130, and a puncturable surface 140. A sachet layer 150 is disposed and substantially sealed about the aperture 130, and an optional envelope layer 190 is also disposed and substantially sealed about the aperture 130 adjacent the sachet layer 150. A reactant 160 is disposed inside vessel 120.

The apparatus 110 also includes a second vessel 220 formed to couple or engage with vessel 120. The second vessel 220 may include a second reactant 265 and the second vessel 220 may be adapted to deliver the second reactant 265 to vessel 120. In the embodiment shown in Figure 2A, the first vessel 120 and the second vessel 220 are configured to engage in a pressure fit configuration as shown in Figure 3A, by applying pressure to the vessels such that the second vessel punctures the first vessel at the puncturable surface. In this embodiment, the puncturable surface is a frangible layer that is ruptured by the first end 221A of the second vessel 220. Once engaged, as shown in Figure 3A, the first vessel 120 and second vessel 220 define the reaction volume and the reactants 160, 265 are contacted. The reaction can then be initiated by initiating agent entering through the sachet layer and/or the initiating agent can be disposed in one of the vessels.

The elements of the apparatus, including the vessels, reactant, sachet layers, aperture, puncturable surface, *etc.*, are as described herein. Preferably, the puncturable surface is constructed from a frangible multi-layer plastic material, *e.g.*, a weak layer of polyolefin positioned near the sealing surface that will fail under pressure. Frangible materials may also include brittle plastic or glass. Suitable frangible materials may be any of the frangible materials described herein or in pending United States Patent Application Serial No. 10/225,769, filed August 22, 2002.

An envelope layer can be included in apparatus of the invention to control or prevent the influx of water vapor into the vessel, while limiting the diffusion of the reactants to the surrounding fluid, be it gaseous or liquid. The envelope layer also allows the gas to diffuse through it to the surrounding fluid from the apparatus. By limiting transmission of initiating agent (*e.g.*, water vapor), into the apparatus, and limiting and/or preventing diffusion of the reactants out of the apparatus of the present invention, the reactant remains concentrated and the pH of the reactive system is localized within the apparatus to optimize the conversion of

reactant to gas. Additionally, intermediates and/or by-products of the reaction, *e.g.*, water, also can contribute to the efficiency and/or duration of the reaction by its affect on the equilibrium of the reactions.

The envelope layer preferably is constructed of a material that is durable and stable. Preferably, it also is capable of fusing to a like material upon the application of heat for construction purposes, *e.g.*, so that two pieces of such material can be fused about its perimeter to an aperture or form an envelope. The envelope layer can be constructed of various materials, including polymeric material, such as perforated films, liquid water permeable films, membranes, and selective transmission films. Suitable envelope layer materials may be any of the envelope materials or envelope layer materials described in pending United States Patent Application Serial No. 10/225,769, filed August 22, 2002, incorporated by reference herein.

At least a portion of the second vessel can be dimensioned to fit within the first vessel, which itself can be formed to receive the second vessel. For example, the vessels can be formed to engage in a pressure fit (*e.g.*, as shown in Figures 2A and 3A), a threaded fit (*e.g.*, as shown in Figures 2B and 3B), a snap fit (not shown), or any known fitting configuration. For example, the vessels can be configured to have a tongue and groove fit or complementary threads that are fit by twisting or pushing the vessels together to force the tongue into the groove or force complementary threads into alignment. After the vessels are coupled, the vessels define the reaction volume, and the reactants and byproducts are substantially retained within the apparatus.

The second vessel may be made from a material having impermeable layers, and contains a compound, *e.g.*, a reactant and/or initiating agent. Optionally, a sachet enclosing a second reactant and/or initiating agent may be disposed within the second vessel. Generally, the second vessel, or at least a portion of the second vessel, is dimensioned to fit within the puncturable surface of the vessel. Like the first vessel, the second vessel may be shaped to suit any particular application, and can have various geometries including cylindrical, oval, spherical, rectangular, and pyramid. The second vessel also preferably has a wall thickness suited to the materials of construction, the reactant and/or the space where the vessel will be placed.

Further, as shown in Figures 2A and 3A, the second vessel wall 222 can be adapted to pierce the puncturable surface 140 of vessel 120 by virtue of the difference in wall heights

224A and 224B. In other embodiments, the wall can form a sharp end and have, *e.g.*, a straight edge or a serrated edge. In another embodiment, the first end features a blunt edge. In yet another embodiment (not shown), the height of the second vessel wall is uniform.

Figure 3A illustrates a method for delivering a gas 210 employing the apparatus 110, described above with reference to Figure 2A. According to this method, the user aligns the first end 221A of the second vessel 220 with the puncturable surface 140 of vessel 120. When the user pushes the first end 221A into the puncturable surface 140, the second vessel 220 creates a hole inor ruptures the puncturable surface 140. The second vessel 220 slides into vessel 120 and couples or engages with vessel 120. In one embodiment, the second vessel 220 slides axially into vessel 120. In this preferred embodiment, the second vessel 220 is engaged with the first vessel 120 by a pressure fit. The second vessel 220 may be adapted to deliver reactant 265 to vessel 120. For example, when the second vessel 220 and vessel 120 couple, the second reactant 265 contained in the second vessel 220 contacts the reactant 160 contained in vessel 120, initiating the reaction to generate the gas 200. The generated gas 200 exits apparatus 110 through sachet layer 150 and optional envelope layer, and gas 210 is delivered to the environment surrounding apparatus 110.

Figure 2B is a cross-sectional side view of another embodiment of an apparatus 110 for delivery of a gas constructed in accordance with the present invention. In general overview, apparatus 110 includes the vessel 120, which defines an aperture 130, and a puncturable surface 140. A sachet layer 150 is disposed about and substantially sealed to the aperture 130. A reactant 160 is disposed insidethe vessel 120. The apparatus 110 also includes a second vessel 220 formed to couple withthe vessel 120. The second vessel may include a second reactant 265 and the second vessel 220 may be adapted to deliver the second reactant 265 to vessel 120. In the embodiment shown, the first vessel 120 is connected with the second vessel 220 in a threaded fit configuration, prior to the rupture of the puncturable surface. Optionally, however, they can be provided separately. The first vessel is formed with the female threads 300 to fit with male threads 310 formed on the second vessel to couple the two vessels by a threaded fit. The second vessel 220 engages the first vessel 120 by actuating the vessels to thread the second vessel into the first, such that puncturable surface 140 is ruptured delivering the reactant 265 contained in the second vessel, as shown in Figure 3B. The second vessel first end 221A is adapted to pierce the puncturable surface 140 of vessel 120.

The elements of the apparatus, including the vessels, reactant, sachet layers, aperture, puncturable surface, *etc.*, are as described herein. Preferably, the puncturable surface is constructed from a frangible multi-layer plastic material, *e.g.*, a weak layer of polyolefin positioned near the sealing surface that will fail under applied pressure. The apparatus can further include additional elements as discussed herein, such as an envelope layer adjacent the sachet layer and sealed about the aperture, a sachet, *etc.*

Figure 3B illustrates a method for delivering a gas 210 employing the apparatus 110, described above with reference to Figure 2B. According to this method, the user aligns the female threads of first vessel 120 and the male threads of second vessel 220 at the first end 221A of the second vessel with the puncturable surface 140 of vessel 120. When the user twists the first end 221A into the puncturable surface 140, the second vessel 220 creates a hole in the puncturable surface 140. The second vessel 220 inserts into vessel 120 and couples with vessel 120. In one embodiment, the second vessel 220 twists axially into vessel 120. In this preferred embodiment, the second vessel is engaged with the first vessel by a threaded fit. The second vessel 220 may be adapted to deliver reactant 265 to vessel 120. For example, when the second vessel 220 and vessel 120 couple, the second reactant 265 contained in the second vessel 220 contacts the reactant 160 contained in vessel 120, initiating the reaction to generate the gas 200. The generated gas 200 exits apparatus 110 through sachet layer 150, and gas 210 is delivered to the environment surrounding apparatus 110.

In another aspect, the invention features a kit for delivery of a gas. The kit generally includes a first vessel defining a reaction volume, the first vessel including a sachet layer disposed about an aperture defined by the vessel, and a puncturable surface. The kit also includes a reactant disposed within the first vessel that generates a gas in the presence of an initiating agent, and a second vessel selectively engageable with the first vessel and formed to provide a compound. The kit can comprise any of the apparatus as described above. The kit optionally can contain initiating agent and/or reactant separately packaged for take up by a second vessel (*e.g.*, a syringe), and delivery to the first vessel. The kit can contain multiple first and second vessels, and the vessels and reactants contained therein can be of different sizes and quantities, so that a user can select appropriate vessels to deliver a desired amount of gas over a desired time period.

In yet another aspect, the present invention features a method of delivering a gas by providing an apparatus for delivery of a gas as described herein, and delivering an initiating agent to the vessel to initiate delivery of a gas. The method can feature any of the steps discussed herein, for example, the method can include the further steps of providing a second vessel formed to engage a vessel of the invention and deliver a compound, and engaging the vessels to the deliver the compound.

The present invention can be used to deliver gas in a wide variety of applications. For example, chlorine dioxide can be used for: the disinfection of water, *e.g.*, municipal water treatment; as a disinfectant for equipment used in the preparation of foods, beverages, fruits and vegetables; for sanitizing fruits and vegetables prior to packaging; for the cleaning and disinfection of medical, dental and food equipment; and for cleaning and disinfecting fluid tanks, fluid lines, and fluid dispensing equipment. Chlorine dioxide has been shown to be an effective disinfectant at concentrations as low as 0.2 mg/L. Chlorine dioxide is a desirable replacement for chlorine, the traditional water treatment chemical, because it has been found to inactivate microbes at lower levels and over a wider pH range. For example, chlorine dioxide can be used to reduce or eliminate biofilms because it penetrates the cell wall of naturally occurring, colony-building microorganisms and disrupts the proteins necessary for reproduction. Moreover, chlorine dioxide does not produce chlorinate by-products, *e.g.*, trihalomethanes. Furthermore, it has been found to be active against pathogens that are resistant to chlorine. It can be used as a slimicide for paper or pulp machines, for wastewater treatment, and for industrial water treatment, *e.g.*, cooling or recycle streams. It can be used for odor control or as an aerial biocide and virucide. It can be used for the treatment of sulfides in the oil industry, for industrial cleaning, *e.g.*, circuit board cleansing, and for paper or tallow bleaching. Sulfur dioxide also has a variety of uses, such as a mold and fungus inhibitor for use in shipping and storing fruits and vegetables. Based on the teachings disclosed herein a practitioner of ordinary skill will appreciate the numerous other applications for which the present invention can be used and provides a heretofore-unmet need.

Where initiation of chlorine dioxide generation is user controlled, and the delivery apparatus controls the release of the chlorine dioxide gas, a predetermined concentration of gas may be supplied to, for example, a confined space. In one embodiment, the apparatus supplies the gas for a specific period of time. A controlled initiation and release of gas into

water to make an aqueous solution may be employed as a biocide fluid flowing through, for example, tanks or fluid lines, for example, in fluid dispensing equipment. In one example, chlorine dioxide gas is dispensed at a predetermined concentration over a specific period of time, such that the user can disinfect a fluid line with chlorine dioxide. After a set time has passed, the user may clear or flush the fluid line of chlorine dioxide thereby taking advantage of the ability of chlorine dioxide gas to quickly "wash out" of fluid lines. Such controlled generation and release of a predetermined gas concentration over a period of time may also be used to disinfect, decontaminate, deodorize or sanitize a confined space, *e.g.*, a postal collection box.

In yet another embodiment, the present invention features a method of manufacturing an apparatus for delivery of a gas. This method comprises the steps of: providing a vessel comprising an aperture defined by the vessel and a puncturable surface; disposing a reactant within the vessel that generates a gas in the presence of an initiating agent; and sealing a sachet layer about the aperture defined by the vessel to define a reaction volume. This method can further include the provision and construction of any of the elements described herein. Further embodiments featuring methods of manufacturing apparatus have been described herein.

Equivalents

Those skilled in the art will recognize, or be able to ascertain using no more than routine experimentation, many equivalents to the specific embodiments of the invention described herein. Such equivalents are intended to be encompassed by the following claims.

What is claimed is:

1. An apparatus for delivery of a gas comprising:

a vessel defining a reaction volume, the vessel comprising:

a sachet layer disposed about an aperture defined by the vessel, and

a puncturable surface; and

a reactant disposed within the vessel that generates a gas in the presence of an initiating agent.

2. The apparatus of claim 1, wherein the reactant comprises an aqueous soluble acid and an aqueous soluble chlorite.

3. The apparatus of claim 1, wherein the sachet layer comprises a hydrophobic material.

4. The apparatus of claim 1, wherein the sachet layer comprises a water vapor selective film.

5. The apparatus of claim 1, further comprising an envelope layer disposed about the aperture and adjacent to the sachet layer.

6. The apparatus of claim 5, wherein the sachet layer comprises a hydrophilic material and the envelope layer comprises a hydrophobic material.

7. The apparatus of claim 1, wherein the apparatus comprises a sachet disposed within the vessel and wherein the reactant is disposed within the sachet.

8. The apparatus of claim 7, wherein the apparatus comprises a second reactant disposed within the vessel.

9. The apparatus of claim 1, wherein the puncturable surface comprises a retractable material or a frangible material.

10. The apparatus of claim 1, further comprising a second vessel formed to engage the vessel and to provide a compound.

11. The apparatus of claim 10, wherein the compound is a second reactant.

12. The apparatus of claim 10, wherein the compound is an initiating agent.

13. The apparatus of claim 10, wherein the second vessel is a needle.

14. The apparatus of claim 10, wherein the second vessel is formed to couple with the vessel and further define the reaction volume.

15. The apparatus of claim 14, wherein the second vessel comprises a male member that is formed to slidably engage the vessel upon actuation of the male member.

16. The apparatus of claim 15, wherein the second vessel and the vessel define complementary surfaces for a snap fit upon actuation of the male member.

17. The apparatus of claim 14, wherein the second vessel and the vessel define complementary threads for threaded engagement of the second vessel and vessel.

18. A kit for delivery of a gas comprising:

the apparatus of claim 1; and

a second vessel selectively engageable with the first vessel and formed to provide a compound.

19. The kit of claim 18, wherein the reactant comprises an aqueous soluble acid and an aqueous soluble chlorite.

20. The kit of claim 18, wherein the sachet layer comprises a hydrophobic material.

21. The kit of claim 18, wherein the sachet layer comprises a water vapor selective film.

22. The kit of claim 18, further comprising an envelope layer disposed about the aperture and disposed adjacent to the sachet layer.

23. The kit of claim 22, wherein the sachet layer comprises a hydrophilic material and the envelope layer comprises a hydrophobic material.

24. The kit of claim 18, wherein the apparatus comprises a sachet disposed within the vessel and wherein the reactant is disposed within the sachet.

25. The kit of claim 24, wherein the apparatus comprises a second reactant disposed within the vessel.

26. The kit of claim 18, wherein the puncturable surface comprises a retractable material or a frangible material.

27. The kit of claim 18, wherein the compound is a second reactant.

28. The kit of claim 18, wherein the compound is an initiating agent.

29. The kit of claim 18, wherein the second vessel is a needle.

30. The kit of claim 18, wherein the second vessel comprises a male member that is formed to slidably engage the first vessel upon actuation of the male member.

31. The kit of claim 30, wherein the first vessel and the second vessel define complementary surfaces to provide a snap fit upon actuation of the second vessel.

32. The kit of claim 18, wherein the first vessel and the second vessel define complementary threads to provide a threaded engagement between the first vessel and the second vessel.

33. A method of delivering a gas comprising:

providing an apparatus for delivery of a gas comprising:

a vessel defining a reaction volume, the vessel comprising a sachet layer disposed about an aperture defined by the vessel, and a puncturable surface, and

a reactant disposed within the vessel that generates a gas in the presence of an initiating agent; and

delivering an initiating agent to the vessel to initiate delivery of a gas.

34. The method of claim 33 further comprising the steps of:

providing a second vessel formed to engage the vessel and deliver a compound; and

engaging the vessel and the second vessel to deliver the compound.

35. A method of manufacturing an apparatus for delivery of a gas, the method comprising the steps of:

providing a vessel comprising an aperture defined by the vessel and a puncturable surface;

disposing a reactant within the vessel that generates a gas in the presence of an initiating agent; and

sealing a sachet layer about the aperture to define a reaction volume.

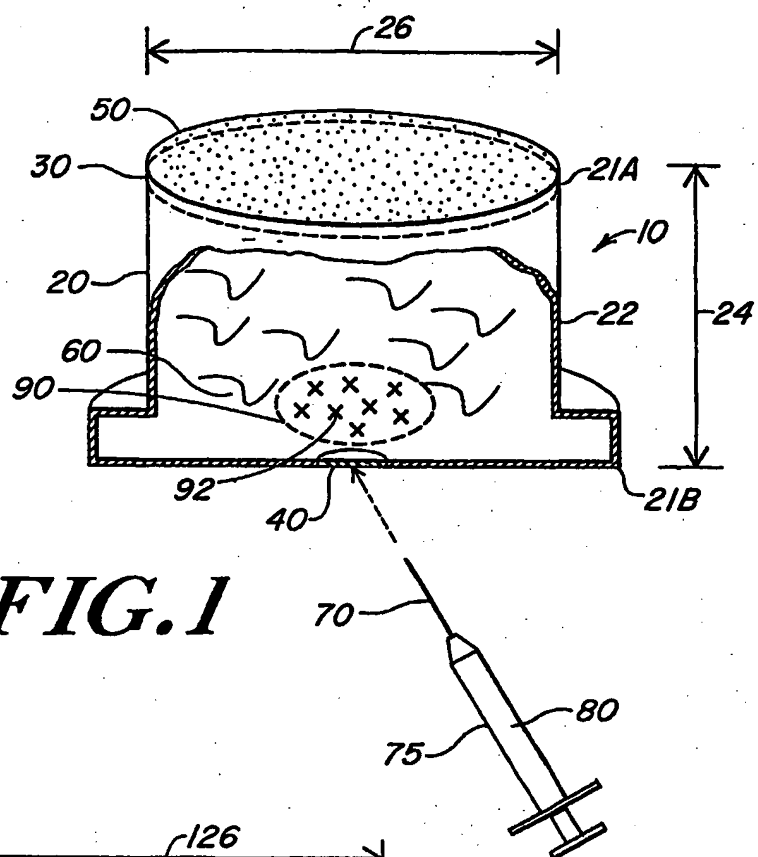
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FIG.1

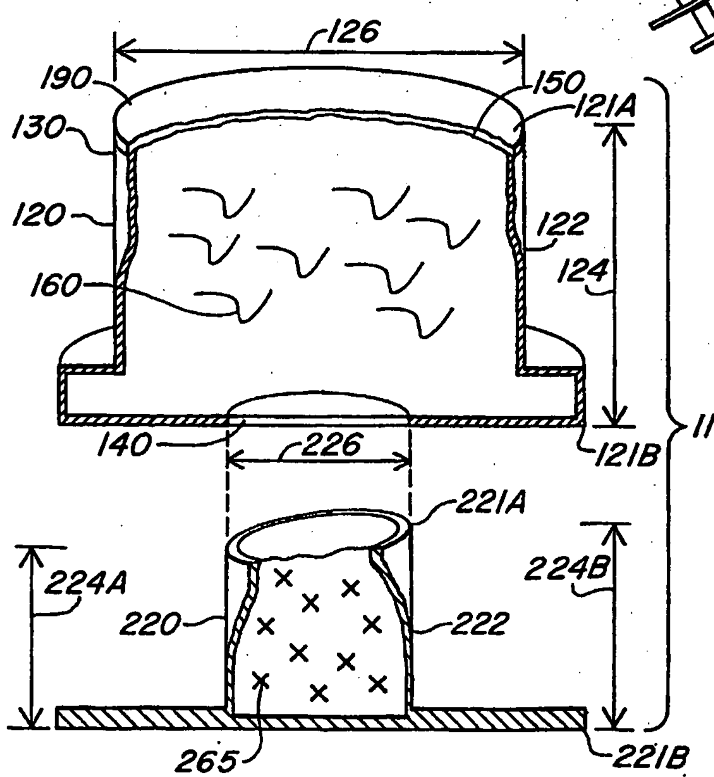
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FIG.2

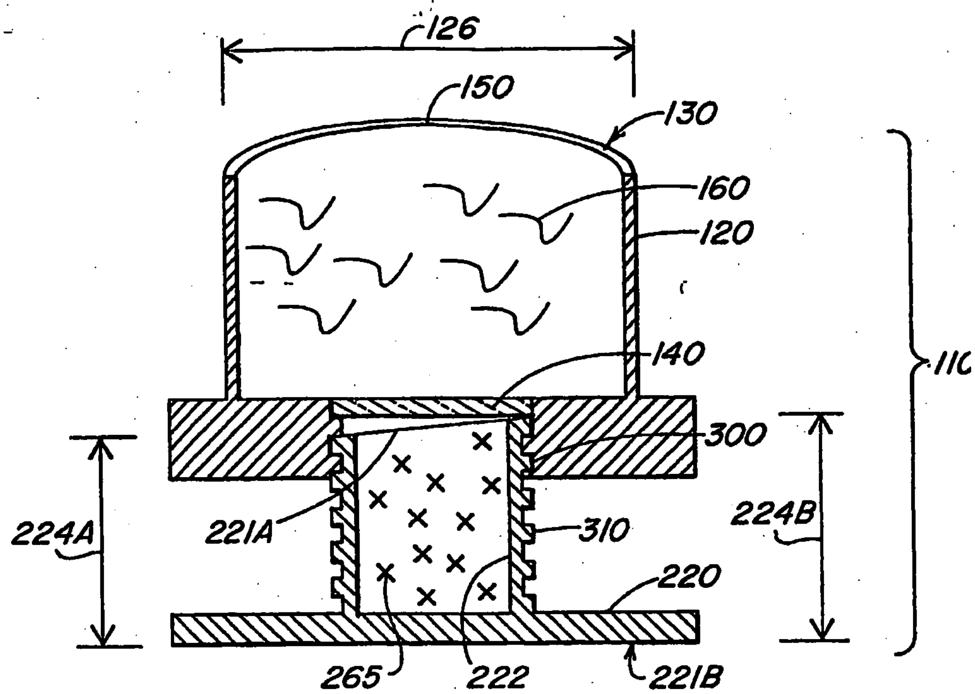
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FIG. 2B

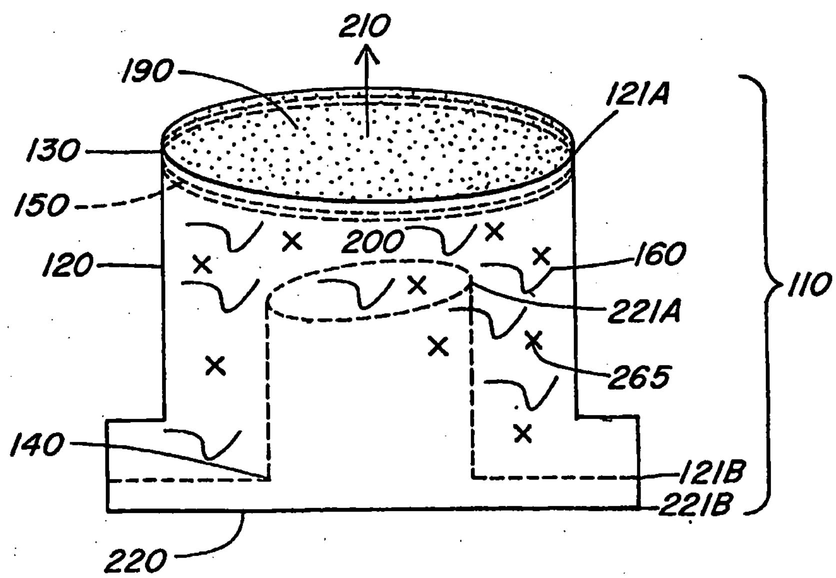
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FIG. 3A

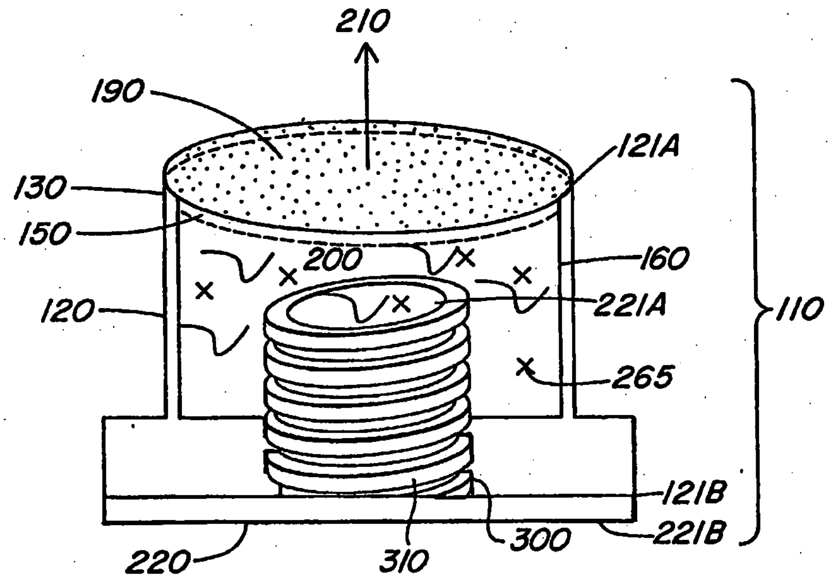
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FIG. 3B

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US02/40302

A. CLASSIFICATION OF SUBJECT MATTER

IPC(7) : A61L 9/00, 2/00; B01J 19/00; A62B 7/08; B01D 11/02; A21D 10/02; A01J 11/04; B65D 25/08
US CL : 422/1, 422/5, 422/28, 422/32, 422/34, 422/36-37, 422/40, 422/123, 422/261, 422/294, 422/300, 422/305

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
U.S. : Please See Continuation Sheet

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched
NONE

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
EAST

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 4,411,918 (Cimino et al.) 25 October 1983, see entire document.	1, 5-16, 18, 22-28, 30-31 and 33-35
Y	US 4,411,918 (Cimino et al.) 25 October 1983, see entire document.	2, 17, 19, 29 and 32.
Y	US 5,705,092 (Wellinghoff et al.) 6 January 1998, see entire document.	3-4 and 20-21

☐ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
07 March 2003 (07.03.2003)	27 MAR 2003

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